

# Strong polaritonic interaction between flux-flow and phonon resonances in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$ intrinsic Josephson junctions: Angular dependence and the alignment procedure.

H. Motzkau, S. O. Katterwe, A. Rydh, and V. M. Krasnov\*

*Department of Physics, Stockholm University, AlbaNova University Center, SE-106 91 Stockholm, Sweden*

(Dated: October 11, 2012)

$\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  single crystals represent natural stacks of atomic scale intrinsic Josephson junctions, formed between metallic  $\text{CuO}_2$ -Ca- $\text{CuO}_2$  and ionic insulating SrO-2BiO-SrO layers. Electrostriction effect in the insulating layers leads to excitation of  $c$ -axis phonons by the ac-Josephson effect. Here we study experimentally the interplay between and velocity matching (Eck) electromagnetic resonances in the flux-flow state of small mesa structures with  $c$ -axis optical phonons. A very strong interaction is reported, which leads to formation of phonon-polaritons with infrared and Raman-active transverse optical phonons. A special focus in this work is made on analysis of the angular dependence of the resonances. We describe an accurate sample alignment procedure that prevents intrusion of Abrikosov vortices in fields up to 17 Tesla, which is essential for achieving high-quality resonances at record high frequencies up to 13 THz.

PACS numbers: 74.72.Hs, 74.78.Fk, 74.50.+r, 85.25.Cp

Intrinsic Josephson junctions (IJJs) [1] in high temperature superconductors are considered as possible candidates for realization of high power THz-frequency radiation sources [2–7]. IJJs can provide several advantages: (i) A large number of IJJs can easily be integrated. A strong superradiant emission can take place if all IJJs are phase-locked in the symmetric in-phase mode. (ii) Due to the large energy gap in Bi-2212  $\Delta \sim 30 - 40$  meV [8], IJJs can comfortably operate in the frequency range up to  $\sim 20$  THz. (iii) Stacking of IJJs may lead to cascade amplification of non-equilibrium population, needed for achieving population inversion and lasing [4]. (iv) A large variety of emission mechanisms are available. The ac-Josephson effect facilitates electromagnetic wave emission via fluxon-induced self-oscillations at geometrical resonances [6, 9], or via the Josephson flux-flow emission [7, 10]. Besides, coherent phonon emission can take place either without the ac-Josephson effect via recombination of non-equilibrium quasiparticles [4], or via the ac-Josephson effect due to the electrostriction phenomenon [11].

In this work we investigate the mutual interaction between phonons and electromagnetic waves in the flux-flow state of small Bi-2212 mesa structures. The ac-Josephson effect leads to excitation of  $c$ -axis optical phonons in Bi-2212 single crystal by means of electrostriction: Ions in the polar insulating SrO-2BiO-SrO layers, where the oscillating electric field is concentrated, are shaken at the Josephson frequency. Both Raman and infrared (IR) - active  $c$ -axis phonons can be excited [11–13]. The symmetry of the phonon is directly related to the symmetry of the electromagnetic mode: Raman phonons are excited by the out-of-phase mode, while infrared-active phonons are excited by the in-phase mode. The strongest interaction between electromagnetic waves and phonons occurs at the transverse optical phonon frequency. This leads to formation of phonon-polaritons with very slow group velocities. We argue that resonances with IR-active phonons can stabilize the in-phase electromagnetic mode in IJJs, needed for achieving coherent superradiant flux-flow emission from Bi-2212 mesas. The appearance of phonon-polaritons also demon-

strates the existence of an unscreened polar response, which may lead to the strong electron-phonon coupling in cuprates.

We study small mesa structures, made at the surface of freshly cleaved Bi-2212 single crystals. Mesas are made by photolithography, argon milling and focused ion beam trimming down to sub-micron sizes. Two types of crystals are studied: slightly underdoped pure Bi-2212 crystals with  $T_c \simeq 82$  K and slightly overdoped lead-doped  $\text{Bi}_{2-y}\text{Pb}_y\text{Sr}_2\text{CaCu}_2\text{O}_{8+x}$  (Bi(Pb)-2212) crystals with  $T_c \simeq 90$  K. The most noticeable difference between those crystals is in the critical current density, which is an order of magnitude larger for Bi(Pb)-2212 ( $J_c \sim 10^4$  A/cm<sup>2</sup>). Since the amplitudes of the phonon resonance, reported below, are proportional to  $J_c^2$  [14], they are much more pronounced in Bi(Pb)-2212 crystals [11]. In order to avoid repetitions we address the reader to our previous works, in which we studied properties of similar mesas in intense magnetic fields, including: the Fraunhofer modulation of the Josephson current [15], velocity matching (Eck), geometrical (Fiske) [10] and phonon-polariton [11] resonances in the flux-flow state, their temperature dependence [16] and the intrinsic tunneling magnetoresistance of IJJs [17]. Below we will focus on the angular dependence and describe the procedure for accurate sample alignment, which is a prerequisite for observation of high quality resonances.

Figure 1 represents the angular dependence of the current-voltage characteristic of the Bi(Pb)-2212 mesa P22/m5b with the area  $\sim 1.3 \times 0.9 \mu\text{m}^2$ , containing  $N \simeq 11$  IJJs. For the field parallel to the  $ab$ -plane, (a)  $\alpha = 0$ , the quasiparticle (QP) branches are completely suppressed and a pronounced flux-flow step appears at  $V \simeq 0.1$  V. It corresponds to the Eck resonance, when the speed of propagating Josephson vortices (fluxons) is equal to the speed of the slowest out-of-phase electromagnetic wave [10, 11, 16]. Strong hysteretic resonances above the Eck step at higher voltages,  $0.1 \text{ V} \lesssim V \lesssim 0.2 \text{ V}$ , are due to excitation of  $c$ -axis optical phonons [11–14, 18]. Already at a slight misalignment of the field by 1-2 degrees (b, c) the resonances are strongly dampened due to

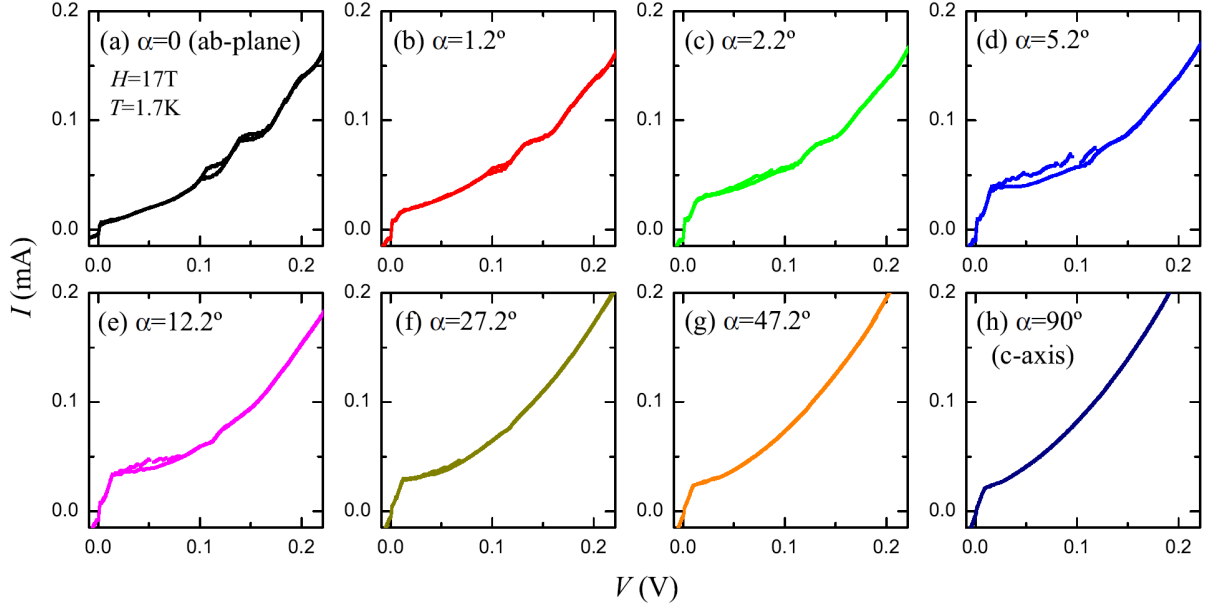


FIG. 1. (Color online). Angular dependence of current-voltage characteristic for a Bi(Pb)-2212 mesa P22/m5b at  $T = 1.7$  K and  $H = 17$  T.

penetration of Abrikosov vortices. The amount of Abrikosov vortices increases with further increase of the angle (d), they effectively pin Josephson fluxons, which leads to a significant increase of the critical current, suppression of the flux-flow and a reappearance of QP branches (d, e). With further increase of the angle the amount of Abrikosov vortices becomes so large that their normal cores start to suppress the superconducting order parameter. This results in the pronounced negative magnetoresistance [17] and the suppression of the critical current (f-h).

Figure 2 represents differential conductance curves  $dI/dV$

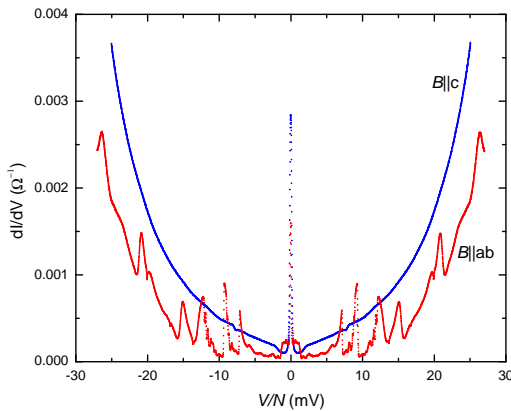


FIG. 2. (Color online). Tunneling conductance versus voltage per junction for a Bi(Pb)-2212 mesa P22/m4a at  $T = 20$  K and  $H = 16$  T in the *ab*-plane and in the *c*-axis direction.

versus voltage per junction  $V/N$  for another Bi(Pb)-2212 mesa (P22/m4a  $1.2 \times 2.0 \mu\text{m}^2$ ,  $N \approx 55$ ) for field along the *ab*-plane and in the *c*-axis direction. Phonon resonances are seen as peaks in  $dI/dV$  for a field along the *ab*-plane. They completely disappear for field in the *c*-axis direction, clearly showing the importance of sample alignment.

Table I summarizes all observed phonon resonances for the Bi(Pb)-2212 crystal. In total fourteen phonon resonances with varying amplitudes could be distinguished. This is consistent with the expected number of *c*-axis optical phonons in the tetragonal Bi-2212 (seven IR and seven Raman) [19]. Indeed, by comparison with optical spectroscopy [19], most of the observed phonons are easily identified with the known Raman and IR-active *c*-axis optical phonons [11–13]. Few unidentified phonons may be disorder induced, due to the orthorhombic distortion, or the incommensurate superstructure in the crystal [19].

From Fig. 1 it is obvious that an accurate sample alignment is essential for the study of phonon resonances in intense fields. Therefore we want to describe the developed unambiguous alignment procedure in more details. Figure 3 shows the angular dependence of the dc-resistances  $R_{dc} = V/I$ , measured at different probe currents  $I$  (a) low  $I = 0.05$  mA, (b) moderate  $I = 0.1$  mA and (c) high  $I = 3$  mA for the Bi(Pb)-2212 mesa P22/m3 at  $T \approx 4$  K and at three different magnetic fields. The angular dependence of the magnetoresistance can be understood from  $I$ - $V$ s in Fig. 1. The sharp maximum at  $\alpha = 0$  is caused by the flux-flow contribution. The flux-flow voltage is rapidly suppressed at small a misalignment  $\alpha \sim 1^\circ$  due to the pinning by Abrikosov vortices. At larger angles the magnetoresistance strongly depends on the bias: at a low bias

$n$	1	2	3	4	5 <sup>a</sup>	6	7 <sup>a</sup>	8 <sup>b</sup>	9 <sup>a</sup>	10	11 <sup>a</sup>	12	13	14 <sup>c</sup>
$V_n$ (mV)	6.0	7.8	10.5	11.2	13.3	14.5	16.0	17.1	19.6	20.6	21.3	23.0	25.7	29.1
type	IR	Raman	IR	Raman	Raman	IR	-	-	Raman	IR	-	IR	Raman	-
symmetry	$A_{2u}$	$A_{1g}$	$A_{2u}$	$A_{1g}$	-	$A_{2u}$	-	-	-	$A_{2u}$	-	$A_{2u}$	$A_{1g}$	-
assignment	Bi:CuCaSr	CuSr	SrCu	Sr:Cu	"disorder"	Ca:Sr	-	-	"disorder"	O3:O1	-	O1:CaO3	O1:Sr	-

<sup>a</sup> very weak

<sup>b</sup> disorder-induced IR-active or  $B_{1g}$  Raman-active

<sup>c</sup> disorder-induced IR-active or  $A_{1g}$  (O3) Raman-active

TABLE I. Experimental voltage positions of phonon resonances. The mean values for all mesas at a Bi(Pb)-2212 crystal P22 are given. Also given are types (Raman-active resp. IR-active), symmetries and assignments of phonons.

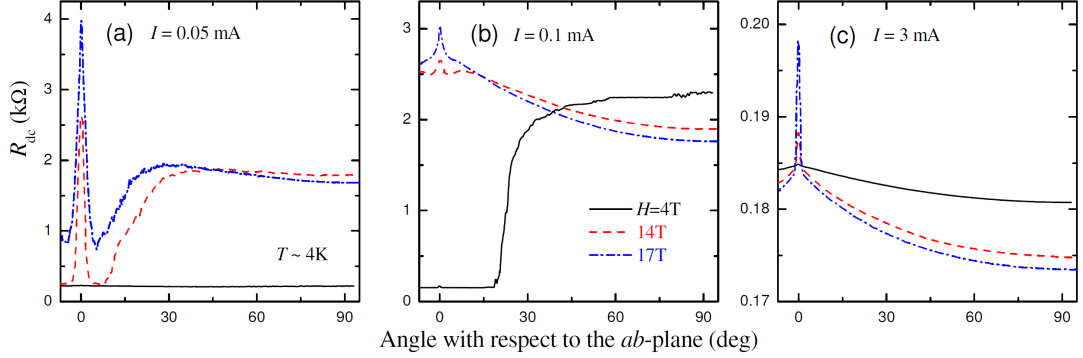


FIG. 3. (Color online). Angular dependence of dc-resistances, measured at different probe currents  $I$  (a) low  $I = 0.05$  mA, (b) moderate  $I = 0.1$  mA and (c) high  $I = 3$  mA for the Bi(Pb)-2212 mesa P22/m3 at  $T \approx 4$  K and at three different magnetic fields 4 T (solid), 14 T (dashed) and 17 T (dashed-dotted line).

the resistance increases due to the suppression of the critical current, but at a higher bias it is monotonously decreasing due to the negative QP magnetoresistance [17, 21].

Even though the sharp flux-flow peak at low bias seems to be advantageous for the sample alignment, we found that such an alignment is unreliable because of the field lock-in along the  $ab$ -plane, which may lead to a hysteresis up to a few degrees [20]. In Fig. 3 (a) it can be seen as a slight asymmetry of the minima in the  $R_{dc}(\alpha)$  curve at  $H = 17$  T. With increasing field the angular range of the hysteresis decreases. At  $H = 14$  T it is only  $\sim 0.2^\circ$ . However this corresponds to almost 500 G  $c$ -axis field that is sufficient for penetration of Abrikosov vortices. Therefore the low bias alignment is not good enough. On the other hand, we observed that the alignment at high bias, as shown in Fig. 3 (c), is working much better because it is free from the hysteresis. This is caused by an effective depinning of Abrikosov (pancake) vortices as a combination of self-heating that leads to thermal-assisted flux creep, a gradient force due to perpendicular magnetization in pancake vortices with respect to applied field and a drag force from Josephson vortices. Furthermore, the maxima  $R_{dc}(\alpha)$  become significantly sharper at high bias. For the curves at  $H = 14$  T the full width at half maximum is  $3.4^\circ$  at low bias, Fig. 3 (a), compared to only  $1.5^\circ$  at high bias, Fig. 3 (c). Therefore we use high bias magnetoresistance for the accurate sample alignment.

Figure 4 shows the measured critical current vs the in-plane magnetic field for (a,b) a moderate size Bi-2212 mesa and (c) a small Bi(Pb)-2212 mesa P22/m4a. A clear Fraunhofer modulation is seen in Figs. 4 (a) and (c). A transition from the half flux quantum to the flux quantum  $\Phi_0$  periodicity occurs with increasing field due to the reconstruction of the Josephson fluxon lattice from the triangular to the rectangular [15, 22]. Panel (b) shows  $I_c(H)$  for the same mesa and alignment as in (a) after subjecting the sample to a larger field  $\sim 8$  T. Apparently, the Fraunhofer modulation is completely destroyed by the intrusion of Abrikosov vortices. In Fig. 4 (c) the Fraunhofer modulation persists up to 13 T, demonstrating that with our rigorous alignment procedure we can prevent Abrikosov vortex intrusion even at very high fields.

Figure 5 shows the  $I$ - $V$  oscillogram for the Bi(Pb)-2212 mesa P22/m5b at  $T = 1.6$  K in the in-plane magnetic field of 10 T. Here a flux-flow branch ending with a nearly vertical Eck resonance is clearly seen. Above it a large variety of phonon resonances appears. Those resonances are divided into groups of  $N = 11$  branches due to one-by-one switching of IJJs into the resonant state. For clarity we painted different families of branches by different colors.

Figure 6 (a) represents the magnetic field dependencies of collective resonance voltages (all junctions at the same resonance) for one of the studied Bi(Pb)-2212 mesas. Positions of all fourteen phonon resonances are indicated by dotted lines.

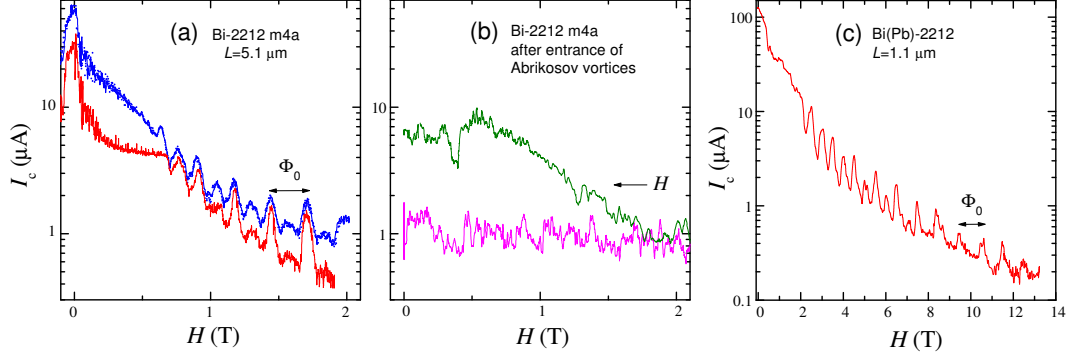


FIG. 4. (Color online). Fraunhofer patterns  $I_c$  vs. in-plane  $H$  for a moderate size Bi-2212 mesa with the length  $L = 5.1 \mu\text{m}$  at  $T = 1.6 \text{ K}$ : (a) For field sweeps from zero to 2.1 T and backwards, (b) After trapping Abrikosov vortices at higher field due to slight misalignment. Different curves in (a,b) represent separate runs. (c) For a small Bi(Pb)-2212 mesa  $L \approx 1.2 \mu\text{m}$  after rigorous alignment at large bias. The clear Fraunhofer modulation indicates the lack of Abrikosov vortex intrusion up to 13 T. Arrows indicate the field for introduction of one flux quantum  $\Phi_0$  in a single junction. A transition from  $\Phi_0/2$  modulation at low fields to  $\Phi_0$  at large fields is due to the reconstruction of the Josephson fluxon lattice from triangular to rectangular.

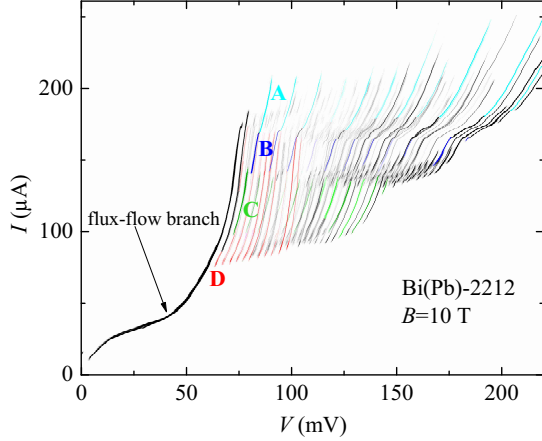


FIG. 5. (Color online)  $I$ - $V$  oscillogram for the Bi(Pb)-2212 mesa P22/m5b at  $H = 10 \text{ T}$  aligned along the  $ab$ -plane. Different families of phonon-polariton resonances are painted in different colors. They are due to one-by-one joining of junctions to the resonant state. The extraordinary large amplitude and hysteresis of the resonances indicates their high quality factors and oscillation amplitudes.

As discussed in Ref. [11], the  $V(H)$  plot can be translated into the dispersion relation with the frequency given by the ac-Josephson relation and the wave length by the periodicity of the Josephson fluxon lattice:

$$f = V/N\Phi_0, \quad (1)$$

$$k = 2\pi Hs/\Phi_0, \quad (2)$$

where  $s = 1.5 \text{ nm}$  is the stacking periodicity of Bi-2212. Therefore the  $V$ - $H$  plot represents the dispersion relation of electromagnetic waves in the mesa. The lowest branch (black) represents the Eck resonance. Initially it increases linearly with field, corresponding to the constant speed of electromag-

netic waves  $\approx 4 \times 10^5 \text{ m/s}$ , close to the out-of-phase speed [10]. However, at  $H > 12 \text{ T}$  it saturates upon approaching the second phonon mode. At this point the group velocity of electromagnetic waves becomes very slow. This is a well known signature for formation of phonon-polaritons with the transverse optical phonon [18].

The polaronic dispersion relation is determined by the dielectric function, which is influenced by the interaction of propagating electromagnetic waves in the dielectric barrier with optical phonons:

$$\varepsilon(\omega) = \varepsilon_\infty + \sum_j \frac{\omega_{\text{TO},j}^2 S_j}{\omega_{\text{TO},j}^2 - \omega^2 - i\gamma_j \omega}. \quad (3)$$

Here  $\omega_{\text{TO},j}$  are frequencies of transverse optical phonons,  $S_j$  and  $\gamma_j$  are the corresponding oscillator strength and the damping parameter, respectively.

Figure 6 (c) shows a simulated dielectric function with characteristic phonon frequencies aligned to the positions of the fourteen observed resonances. The oscillator strengths were chosen so that  $\varepsilon(0) = \varepsilon_\infty + \sum_j S_j$  is equal to the experimental value  $\sim 10$  and damping parameters were chosen so that the corresponding dispersion relation  $\omega = c(0)k/\sqrt{\varepsilon(\omega)}$ , shown in Fig. 6 (b) is looking alike the experimental data in Fig. 6 (a). Figure 6 (b) shows the obtained dispersion relation for positive group velocities with scales in frequency and the wave number corresponding to voltage and field scales in Fig. 6 (a) according to Eqs. (1) and (2). Even though Fig. 6 (b) is not meant to be a direct fit to Fig. 6 (a), it demonstrates that peculiarities of experimentally observed  $V(H)$  characteristics can be fairly well explained by the simple expression Eq. (3). In particular, the saturation of  $V(H)$  and small group velocities are caused by the divergence of the dielectric function when the ac-Josephson frequency is approaching any of the

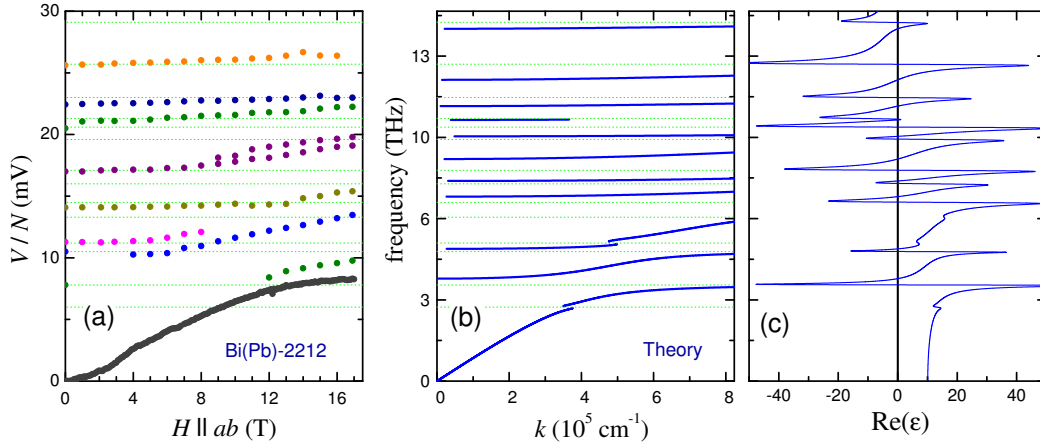


FIG. 6. (Color online). (a) Magnetic field dependence of voltages (per junction) of different resonances for one of the Bi(Pb)-2212 mesas. It can be translated into the dispersion relation  $f(k)$  according to Eqs. (1) and (2), which correspond to the scales in panel (b). Positions of fourteen characteristic phonon frequencies are marked by dotted lines. Panels (b) and (c) represent the calculated dispersion relation and the corresponding polaronic dielectric function, given by Eq. (3).

active transverse-optical phonon frequencies. Also an abrupt jump-like cancellation of resonances at higher voltages (see Fig. 5) is caused by the negative value of  $\epsilon$ , which prevents propagation of electromagnetic waves with such frequencies and leads to switching of  $I$ - $V$ s from the resonant flux-flow to the non-resonant quasiparticle state.

In conclusion, we have described the accurate alignment procedure and the angular dependence of phonon-polariton resonances between electromagnetic waves, generated in the flux-flow state, and  $c$ -axis optical phonons in Bi-2212 single crystals. The phenomenon is caused by electrostriction in the insulating SrO-2BiO-SrO layers. Our work has several implications. For realization of the coherent flux-flow oscillator it is necessary to stabilize the in-phase state with the rectangular Josephson fluxon lattice, which is usually unstable because of fluxon-fluxon repulsion. Strong interaction with infrared optical phonons can stabilize the in-phase state. The stabilizing role of phonons is seen already from the fact that we observed the flux-flow phenomenon up to the record high frequencies  $\sim 13$  THz, see Fig. 6 (a) and (b). Formation of phonon-polaritons demonstrates also that a residual polar response is left in cuprates even in the superconducting state. This demonstrates that Bi-2212 single crystals represent natural atomic metamaterials composed of two-dimensional superconducting  $\text{CuO}_2$ -Ca- $\text{CuO}_2$  layers stacked with polar insulating SrO-2BiO-SrO. This can lead to an extraordinary strong electron-phonon coupling due to the combination of a long range Coulomb interaction in 2D metallic planes with an unscreened polar response in the adjacent polar insulator layers. This may have significance for understanding the pairing mechanism in cuprate superconductors.

Financial and technical support from the Swedish Research Council and the SU-Core Facility in Nanotechnology is grate-

fully acknowledged.

\* Vladimir.Krasnov@fysik.su.se

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